

AAS021-01

Room:102

Time:May 23 08:30-08:45

Carbon budget estimation by inverse modeling with atmospheric CO₂ concentrations from surface and CONTRAIL measurements

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A great deal of understanding of the global and regional carbon budget helps us to perform a reliable prediction of future climate with an earth system model. However, the accuracy of CO₂ source/sink estimation by inverse modeling, which is one of the leading methods to estimate regional carbon budget, is not very high because of sparse observational data coverage. The recent evolving aircraft measurements of CO₂ in a three-dimensional view are expected to provide new constraints on the estimation of surface CO₂ fluxes.

In this study, regionally divided carbon budgets are estimated by inverse modeling using surface measurement networks and aircraft measurements from Comprehensive Observation Network for Trace gases by Airliner (CONTRAIL). The CONTRAIL project has started since late 2005 and a huge amount of atmospheric CO₂ data has been obtained covering altitudes between the Earth surface to the upper-troposphere and lower-stratosphere, latitudes between the boreal high-latitudes to the austral mid-latitudes. Monthly mean observational data from GLOBALVIEW-CO₂, which mostly consists of surface measurements, and CONTRAIL are used in this inverse analysis. The CONTRAIL data measured both vertically over each airport and horizontally at the cruising altitude are used. The inversion method is based on the Bayesian statistics and the approach of the TransCom 3 is used. To relate atmospheric concentrations to surface fluxes, a three-dimensional transport model is employed. In this study, Nonhydrostatic ICosahedral Atmosphere Model (NICAM)-based transport model (NICAM-TM) is used with prescribed CO₂ flux data of fossil fuel emission, respiration/photosynthesis in terrestrial biosphere and atmosphere-ocean exchange. The transport simulations of atmospheric CO₂ are performed by low-resolution version of NICAM; the horizontal grid interval is about 240 km. The analyzed period is five years during 2005-2009 and meteorological fields in each year are used to drive the transport model with the nudging method. The inversion setup is similar to TransCom3, but number of flux regions to be estimated is 42. Land regions are divided into 31 according to vegetation types and the same 11 ocean regions as TransCom 3 are used.

A preliminary result of forward simulation with the prescribed fluxes shows that the transport model has good performance for reproducing general features of three-dimensional structure of CO₂ observed by CONTRAIL. However, some discrepancies between the simulation and CONTRAIL are found in horizontal gradient even in the upper-troposphere during summer, indicating that aircraft measurements have significant impacts on flux estimates when vertical transport is efficient. The inversion results will be discussed in the presentation.

Keywords: carbon cycle, inversion

AAS021-02

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CO₂ seasonal distributions in the UT/LS region as observed by CONTRAIL and four transport models

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In the Comprehensive Observation Network for Trace gases by AirLiner (CONTRAIL) project, high-frequency and wide-ranging CO₂ data in the upper troposphere(UT)/lower stratosphere(LS) region have been obtained by Continuous CO₂ Measuring Equipment (CME) onboard commercial aircraft operated by Japan Airlines (JAL).

The observed distributions in UT/LS region showed that CO₂ isopleths followed the tropopause during the winter and spring. On the other hand, distributions tracked potential temperature surfaces crossing the tropopause in summer, suggesting fast meridional transport of high CO₂ from the tropical troposphere. However, it is difficult to show how the tropospheric air masses intrude into the lower stratosphere across the tropopause due to the limited observational data.

Recently, we conducted CONTRAIL transport model intercomparison to improve our knowledge of three-dimensional structures of atmospheric CO₂. The distributions in UT/LS region simulated by four global chemical transport models (ACTM, MJ98-CDTM, NICAM-TM, NIES) with common CO₂ flux dataset were used to study the transport processes near the tropopause. It was shown that the models reproduced observed CO₂ distributions following the tropopause in winter-spring season, although the CO₂ gradients across the tropopause are underestimated by approximately 2 ppm between 300 and 350 K in potential temperature surfaces. In summer, the isentropic transport of high CO₂ from the upper-troposphere in lower latitudes to the lower-stratosphere in higher latitudes were well simulated by the models. The detailed processes of transport will be analyzed and shown in the meeting.

Keywords: CO₂, transport model, UT/LS exchange

AAS021-03

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JMA aircraft observation for greenhouse gases using a cargo aircraft C-130H to Minamitorishima

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Japan Meteorological Agency (JMA) started a long-term aircraft observation of greenhouse gases as one of the operational atmospheric monitoring activities in 2011. In cooperation with the Ministry of Defense, a cargo aircraft C-130H is used for the flask sampling observation during a roundtrip flight to Minamitorishima (MNM) once a month. The air samples are collected during a cruising flight at about 6 km as well as a descending to MNM. After the flight, we measure 4 trace gas concentrations of carbon dioxide (CO₂), methane (CH₄), carbon monoxide (CO), and nitrous oxide (N₂O). Before the regular observations, preliminary observation flights using the C-130H were made in 2010 to evaluate a new flask sampling equipment and a high-precision measuring system with the collaboration of Meteorological Research Institute (MRI). In this presentation, the details of newly developed instrumentations for JMA aircraft observation and their performances are introduced.

Air samples are pressurized into the flasks by a manual diaphragm pump to an absolute pressure of about 0.4MPa. To minimize the drifts of trace gas concentrations in the flasks, we specially prepared a 1.7-L titanium flask of which internal surface is coated by silica. The storage tests for the flask samples during several days were repeated to ensure the stability of trace gases until analyses. To avoid the contamination of cabin air, sample air was taken from an air-conditioning blowing nozzle upstream of the recirculation fan. Specially coordinated flights at a low altitude of 1000ft over MNM were made using the C-130H to compare with the ground-based measurements from the MNM monitoring system operated by JMA. From these comparison experiments, it was confirmed that our aircraft sampling procedure was suitable for the precise measurements of trace gases.

JMA/MRI developed a new automated measuring system consisting of a conventional NDIR analyzer (Licor, LI-7000) for CO₂ as well as recently advanced spectroscopy instruments of WS-CRDS analyzer (Picarro, G2301) for CH₄, VURF analyzer (Aero-Laser, AL5002-AIR) for CO, and off-axis ICOS analyzer (Los Gatos, N₂O/CO Analyzer) for N₂O. A lot of test runs using standard gases and natural air indicated that higher-precision analyses could be easily achieved rather than before, instead of the complicated GC systems, although relatively larger amount of sample air is required.

Keywords: aircraft observation, greenhouse gas

AAS021-04

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Evaluation of CO₂ emission from the Tokyo metropolitan area based on balloon born measurements: Simultaneous observation

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Emissions of CO₂ have increased drastically over the past century as a result of the mass consumption of fossil fuels due to the expansion of industrial activities resulting in dramatic increases in atmospheric concentrations of CO₂. CO₂ emissions from urban area are an important term of global carbon budget, but its estimation is mainly based on inventories of fossil fuel consumption and road traffic. To ascertain the CO₂ flux values from urban area, detailed CO₂ measurements including vertical distribution are required.

In this study, simultaneous measurements of CO₂ vertical profiles using originally developed balloon borne instruments (CO₂-sonde) at three sites on January 7, 2011 to evaluate CO₂ emission from the Tokyo metropolitan area. The three sites, Isezaki (Gunma Pref.), Ichihara (Chiba Pref.), and Shirako (Chiba Pref.), where the balloon borne instruments were launched, are located upwind, inside, and downwind of the metropolitan area, respectively. The CO₂ sensors are based on a non-dispersed infrared absorption spectroscopy technique at the wavelength around 4.3 micrometers. The data of the optical infrared absorption are transmitted through a GPS rawin-sonde (Meisei RS-06G) with temperature, humidity and GPS data every second.

As a result, at lower altitude (<1 km), CO₂ mixing ratio obtained at Ichihara are higher than those obtained at other sites by 2-7 ppmv, while the three vertical profiles are indistinguishable at free troposphere. These observational data will be used to evaluate CO₂ emission from the Tokyo metropolitan area using CO₂ transport models and also to validate CO₂ total column measurements by the greenhouse gas observing satellite (GOSAT) and a ground base fiber-etalon spectrometer.

Keywords: Carbon dioxide emission, Balloon-borne measurement, Tokyo metropolitan area, Satellite validation

AAS021-05

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Real-time measurements of CO₂ stable carbon isotope ratio in the atmosphere using wavelength modulation spectroscopy

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1. Introduction

Measurements of the stable isotopes provide important information on the source and history of their compounds in many research areas. Since the isotope ratio changes in relation to the conditions, the real-time measurements are required. It is recognized that the standard measurement technique for the stable isotope ratio is isotope ratio mass spectroscopy (IRMS). The IRMS has high precision in the range from 0.01 to 0.1 per mill. Nevertheless, pre-treatment is needed when a gaseous sample is injected in the mass spectrometer. Therefore, it is difficult to perform the real-time measurements of the isotope ratio. Recently, the laser absorption spectroscopy has been applied to the measurements of the stable isotopes. The isotopomers are easily recognized without interference of other species, when the absorption line is selected appropriately. Using this technique, since the sample gas is just introduced into the sample gas cell without any pre-treatment, the real-time measurements are able to be performed. Several studies using a quantum cascade laser with the direct absorption technique and a DFB laser with wavelength modulation spectroscopy or cavity ring-down spectroscopy have been performed. However, the precision is insufficient. Therefore, in this paper, higher precision measurements of the CO₂ stable carbon isotopes have been performed using WMS with a 2008 nm DFB laser.

2. Experimental

A 2008-nm single-mode DFB laser diode with a typical output power of 10 mW was used as a light source. The beam was introduced into a wedged-window Herriott-type multi-pass cell. The cell had a path length of 29.91 m and a volume of 0.9 L. The transmitted laser beam was focused using an AR-coated CaF₂ lens on an InGaAs photodiode detector. The wavelength of the DFB laser was modulated sinusoidally at 11 kHz using the output of a digital lock-in amplifier. The laser was scanned at 0.77 Hz by changing the injection current, which was controlled using a function generator that supplied a triangle voltage wave. The detected line pairs were ¹²CO₂ at 4978.205 cm⁻¹ and ¹³CO₂ at 4978.023 cm⁻¹. The stable isotope ratio was determined with comparing the measurements ¹³C/¹²C ratio to the international PDB-standard ¹³C/¹²C. All experiments were performed at 313 K. The pressure in the cell is kept 10 kPa.

3. Results and Discussion

The second-harmonic WMS spectra of the sample CO₂ gas, 379 ppm and -30.5 per mill, are detected. The S/N (Signal to noise ratio) of ¹³CO₂ is approximately 66, even though the concentration of ¹³CO₂ is 100 times lower than that of ¹²CO₂. It was found that this line pair is favorable because of their clearly separated signals and comparable intensities. The continuous stable carbon isotope ratio measurements were performed using this apparatus to investigate long-term stability. The signal was measured during 10 hours with 5 minutes increment. The pressure and temperature were found to be controlled within 0.01 kPa and 0.1 K, respectively, during continuous measurements. In this long-term measurement, the precision was achieved to be 0.1 per mill. Then, the continuous stable carbon isotope ratio measurements in ambient air was performed. The CO₂ concentration was also measured with NDIR. The CO₂ concentration profile obtained by our constructed apparatus was good agreement with that obtained with NDIR. The stable carbon isotope ratio of CO₂ was also measured successfully using this system. It was found that its value decreased with increasing CO₂ concentration.

4. Conclusions

The stable carbon isotope ratios of CO₂ were measured using WMS with a 2008 nm DFB laser diode. The precision was achieved to be 0.1 per mill for long-term detection (10 h). The stable carbon isotope ratios of CO₂ was successfully measured using our constructed system.

Keywords: Measurements of carbon isotope ratio, Carbon dioxide, Wavelength modulation spectroscopy, Multi-pass cell, DFB laser

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AAS021-06

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The vertical profiles of CH₄ observed at Tsukuba with a Fourier transform spectrometer

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Fourier transform spectrometer (FTS) has advantages in its high resolution and the wide wavenumber range. Vertical profiles of some species can be derived from the high-resolution spectra. The vertical profiles and column densities of CH₄ were retrieved from the solar spectra observed at Tsukuba, Japan with SFIT2 spectral fitting program developed by Rinsland et al. (1998). It needs to select an appropriate wavenumber region and the optimization of fitting parameters are also needed. Now we are investigating these parameters in the NDACC/IRWG group and we will reanalyze the vertical profiles and column densities of CH₄. We found that the phase of seasonal variation of the mixing ratios in the lower stratosphere is shifted from those in the troposphere and the temporal variation of total column shows steplike increase in 2007 from preliminary analysis.

Keywords: FTIR, Trace Species, Methane

AAS021-07

Room:102

Time:May 23 10:00-10:15

Long-term trend of methane concentration in the lower stratosphere over Japan

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Stratospheric methane acts as an important source of stratospheric water vapor and also a sink for chlorine radical. For predicting the future of the stratospheric chemistry and dynamics, as well as for evaluating strategies for limiting or reducing future emission of methane into the atmosphere, it is indispensable to figure out the past methane trend in both of stratosphere and troposphere accurately. Systematic collections of stratospheric air samples have been carried out over Japan since 1985, using a balloon-borne cryogenic sampler. The air samples collected were analyzed for the CH₄ and N₂O concentrations since 1988. Almost linear and compact relationship between CH₄ and N₂O concentrations was found for all observations. It is well known that tropospheric N₂O has been secularly increasing quite monotonously in recent decades. On the other hand, increase rate of tropospheric CH₄ has been varied complicatedly. Despite the fact that both concentrations have been increased independently in troposphere, there is no significant difference between the correlations of stratospheric CH₄ and N₂O concentrations in each year, at first glance. Considering this fact, the compact relationship obtained over Japan suggests that both CH₄ and N₂O are destroyed at similar rate in the lower stratosphere during the poleward transport of stratospheric air, although the chemical destruction processes of the two gases are quite different. Therefore, we employed N₂O-depressions, instead of N₂O concentrations, for examining correlations with CH₄ concentrations. This method cancels the effects of secular N₂O increase in stratosphere and enables us to detect possible change in stratospheric CH₄. Thus we found that stratospheric CH₄ shows a significant increase before 2000 and clear stagnation after 2000. We categorized CH₄ concentration data into different N₂O depression ranges, and calculated increase rates by applying the curve fitting procedure, taking into account the age of stratospheric air. Average increase rates were calculated to be about 0.4 and 0.1%/year before and after 2000, respectively, in the lower stratosphere. This rate is comparable with the results of 207 ppbv increase in the period of 1978 to 2003 in the lower stratosphere reported by Rohs et al.(2006). However, increase rate before 2000 obtained in this study is much smaller than the result of 1.95 and 0.87 %/year in 1985 and 1994, respectively, by ATMOS/ACE-FTS measurements (Rinsland et al., 2009).

Keywords: methane, stratosphere, trend

AAS021-08

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Time:May 23 10:15-10:30

Latitudinal distribution of APO seasonal cycles and its relation to the meridional circulation in the lower troposphere

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We examine the latitudinal differences in the phasing of the average seasonal cycles of the atmospheric potential oxygen ($APO = O_2 + 1.1 \times CO_2$), which is based on the atmospheric O_2 and CO_2 observations in the western Pacific region. Because APO is invariable with respect to the terrestrial biotic exchanges, its seasonal variations mainly reflect air-sea exchanges of O_2 . Investigating on the APO gives new insights into the meridional circulation because APO is a tracer from the ocean, which has very different flux distribution from those of land tracers. The seasonal minimum occurs in March and September in the Northern and Southern Hemispheres, respectively, and the latitudinal distribution of the date of the seasonal minimum shows discontinuous change at the equator. Contrary to this, the date of the seasonal maximum smoothly changes across the equator from March at 35 deg. S to July at 15 deg. N and levels off between 15 deg. N and 50 deg. N. The seasonal variation in APO is predominantly driven by the air-sea O_2 fluxes between 30-60 deg. in both hemispheres because seasonal variation in the air-sea O_2 fluxes in the equatorial regions are relatively small. Therefore, the seasonal cycles of APO in the tropics depend mainly on the meridional propagation of the atmospheric signals in the lower troposphere. The observational results indicate that the seasonal minima propagate equatorward with little phase lags in both hemispheres and the seasonal maximum propagate equatorward with a substantial phase lag in the Southern Hemisphere. These seasonal differences in the propagation speed could be explained by the strong and weak meridional circulation in winter and summer, respectively, in the lower troposphere. The latitudinal distribution of the date of the seasonal maximum in the Northern Hemisphere may be attributed to the influence of the propagation of the seasonal maximum in the Southern Hemisphere because the seasonal variations in the air-sea O_2 fluxes is about 2 times larger in the Southern Hemisphere than in the Northern Hemisphere.

Keywords: APO, seasonal cycle, meridional circulation, atmospheric oxygen, air-sea gas exchange

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AAS021-09

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14C measurements in suburban aerosols from northern Japan: An enhanced production of biogenic organic aerosols in spring

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Here, we report the year-round observation of fossil and modern carbon in total carbon (TC) and water-soluble organic carbon (WSOC) in atmospheric aerosols from Sapporo, northern Japan, based on radiocarbon measurements. We found that modern carbons are more important in both TC and WSOC, except for wintertime, with elevated levels in spring. Interestingly, WSOC showed higher percent modern carbon (pMC) throughout the year, suggesting that WSOC is produced by photochemical oxidation of biogenic volatile organic compounds especially during spring to summer.

Keywords: aerosols, radiocarbon, total carbon, water-soluble organic carbon

AAS021-10

Room:102

Time:May 23 11:00-11:15

Significant contribution of isoprene oxidation products in summertime organic aerosols at the summit of Mt. Fuji, Japan

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We investigated the organic molecular compositions and size distributions of the summertime aerosols collected at the summit of Mt. Fuji (3776 m, a.s.l.) using gas chromatography/mass spectrometry. More than 120 organic species were detected in the aerosols and grouped into different compound classes such as aliphatic lipids, sugars, phthalate esters, sterols, hydroxy-/polyacids, and biogenic secondary organic aerosol (SOA) tracers for the photooxidation of isoprene (e.g., 2-methyltetrols), alpha/beta-pinene (e.g., pinic acid), and beta-caryophyllene (beta-caryophyllinic acid). Total concentrations of the identified organics were 76.1-325 ng m⁻³ (average 183 ng m⁻³) for the whole-day samples, which are more than 10 times higher than those in nighttime samples (9.28-19.2 ng m⁻³, average 15.6 ng m⁻³). Higher concentrations of both primary and secondary organic marker compounds were observed in the whole-day samples, indicating that the mountain venting at Mt. Fuji should act as an efficient pump that uplifts the ground-surface aerosols and their precursors to the free troposphere. Interestingly, isoprene SOA tracers (2.87 ng m⁻³ in nighttime and 69.2 ng m⁻³ in whole day) were found to be the most abundant compound class. 2-Methylerythritol and 2-methylthreitol, the well-known isoprene SOA tracers, were detected as the dominant single compounds. Using a tracer-based method, we estimated the concentrations of secondary organic carbon (SOC) derived from isoprene, alpha/beta-pinene, and beta-caryophyllene to be 2.16-51.2 ngC m⁻³ (15.5 ngC m⁻³) during the nighttime and 183-954 ngC m⁻³ (465 ngC m⁻³) during the whole day. These values correspond to 0.80-31.8% (12.5%) and 21.6-48.9% (31.9%) of the organic carbon (OC) concentrations in nighttime and the whole-day samples, in which isoprene-derived SOC accounts for 80% and 72% of total SOC, respectively. This indicates that a large amount of organic aerosols in the free troposphere should be derived from the oxidation of isoprene emitted from the forest areas on the foothills. Size distributions of the identified organics were unimodal in most cases. Biogenic SOA tracers (e.g., 2-methyltetrols and 3-hydroxyglutaric acid), levoglucosan and malic acid were detected in the fine mode, while sucrose and trehalose that are abundant in airborne pollen and dust aerosols peaked in the coarse mode. This study provides useful information to understand the sources and abundances of organic aerosols over high mountains in East Asia.

Keywords: organic aerosols, Mt. Fuji, levoglucosan, 2-methyltetrols, biogenic VOCs, isoprene

AAS021-11

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A field experiment to determine the CCN activity of primary marine aerosols generated from natural seawater

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Oceanic surface waters contain a large amount of organic substances produced by marine biota, which are transferred to the atmosphere as primary marine aerosols (PMA) by bubble bursting processes. The organics potentially play an important role in regulating the physico-chemical properties of the PMA, including the cloud condensation nuclei (CCN) activity. Our current knowledge on the relationship of the organics in PMA and the particle properties is, however, still limited despite recent laboratory/field studies on PMA. To better characterize the CCN activity of PMA associated with the organics, we performed an experiment of particle production by the bubble bursting of natural seawater.

The experiment was conducted in Maizuru Bay, the Japan Sea, onboard R/V Ryokuyo-Mar, Maizuru Fishery Research Station, Kyoto University. The PMA generator, which floats on the sea, was prepared to produce aerosols by the bursting of air bubbles in natural seawater. The generator is equipped with a bubble-producing glass ball filter, through which compressed dry air was passed, at 30 to 40 cm below the air-sea interface. The bubbles rose to the sea surface and burst inside a 30 L PTFE dome. The generated PMA were transferred to the instruments onboard through PTFE tubing. Dried PMA was introduced to a differential mobility analyzer (DMA) for size selection, and the resulting monodisperse aerosol was transferred to a condensation particle counter and a continuous flow thermal gradient CCN counter to measure the number concentrations of condensation nuclei (CN) and CCN, respectively. The activation diameters (D_{act}) of the PMA at supersaturations ranging from 0.1% to 0.5% were calculated from the CCN to CN ratios. Surface seawater samples were also collected, which were used for the determination of chlorophyll-a (chl-a) concentrations.

The chl-a concentrations inside Maizuru Bay (IMB) were much higher than that outside Maizuru Bay (OMB). The D_{act} of the generated PMA at IMB and OMB were clearly different in particular at higher supersaturations (0.3% and 0.5%); D_{act} of PMA at IMB were larger than that of PMA at OMB. The hygroscopicity parameter kappa (Peters and Kreidenweis, 2007) calculated for the PMA at the highest supersaturation were lower than that of sodium chloride, suggesting that the PMA were the mixture of less hygroscopic organics and sea salts. The results suggest that organics in seawater transferred to the atmosphere as PMA are enriched especially in the ultrafine mode and affect the particle CCN activity.

Reference: Petters, M. D., and S. M. Kreidenweis (2007), A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, *Atmos. Chem. Phys.*, 7(8), 1961-1971.

Keywords: primary marine aerosols, cloud condensation nuclei, organics, natural seawater

AAS021-12

Room:102

Time:May 23 11:30-11:45

Intensive field observations of trace gases/aerosols in Rudong, China in spring 2010: Objectives and initial results

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Although Central East China is regarded as an emerging region with high emission rates of precursors of ozone and aerosols, available observational data are still limited. We conducted an intensive field campaign observing ozone and its precursors and chemical components/physical and optical parameters of aerosol particles at Rudong (32.26N, 121.37E), Jiangsu, China in May/June 2010 under international collaboration. The location is near the west side of Yellow sea and is away from Shanghai by 100 km and from Rudong city center by 15 km. In this presentation, objectives of the field measurement and initial results are summarized. The main objectives are to characterize typical emission ratios of primary pollutants for this region (including those from biomass burning), to examine the mass closure and point optical closure of aerosols and to obtain useful information to evaluate and improve the capability of the tropospheric chemical transport model to simulate PM_{2.5} and AOD, to study ozone production rates/controlling factors including the effect of aerosols on ozone photochemistry, and to achieve instrument intercomparisons to improve our observational capabilities. Three distinct periods are clearly found: relatively polluted period (possibly under influence from urban, May 15-28), clean period with easterly wind (May 29? June 9), and post-harvest biomass burning period (June 10- 24). The BC/CO ratio during the biomass burning period was clearly higher than other periods, in accordance with the emission inventory. The features of the BC/CO and other ratios potentially characteristic to emission sources are compared to past observations in China. Within the 1.5 month observation period, we found several cases where the air mass traveled from Rudong to Fukue Island (32.75N, 128.68E), located at the opposite east side of the Yellow Sea, allowing the direct comparison of the observational features at the two sites. At Rudong, photolysis frequencies (J values) important to ozone photochemistry were found to be attenuated significantly by the presence of dense aerosols. The effect of the aerosols on ozone photochemistry was analyzed. The relationship between the increase in the aerosol scattering coefficient with relative humidity and chemical composition of aerosols was analyzed and the results were compared to those for Fukue Island in the previous year. Comparison of black carbon concentrations measured by three different instruments resulted in a magnitude relationship similar to those obtained at different locations (e.g., Fukue Island and Mt. Tai), contributing to reduction of the observational uncertainty or to improvement of the consistency among the existing black carbon concentration data observed by different instruments in East Asia.

Keywords: China, ozone, aerosol, intensive field observation

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AAS021-13

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PM_{2.5} variation in Rudong, China and Fukue Island, Japan in spring 2010

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We carried out an intensive field campaign observing ozone and its precursors and chemical components/physical and optical parameters of aerosol particles at Rudong (32.26N, 121.37E), Jiangsu, China in May/June 2010. In this presentation, to investigate the PM_{2.5} mass concentration, their composition and correlation in both Rudong and Fukue Island(32.75N, 128.68E), we observed PM_{2.5} mass concentration using SHARP monitor and sampled PM_{2.5} on the quartz filter by high volume air sampler. PM_{2.5} total mass concentration is monitored every 1 min for both sites. At the Rudong site(May16-June23), 9 or 14-hours PM_{2.5} samples were collected on the quartz filters using High-volume(500L/min) sampler, while 22-hours PM_{2.5} samples were collected at the Fukue Island site(May18-25). In the end of May, high PM_{2.5} mass concentrations were observed in both Rudong and Fukue Island. We are going to discuss mass closure and correlation of the PM_{2.5} for Rudong and Fukue Island results.

Keywords: PM_{2.5}, aerosol, composition, transportation, mass closure

AAS021-14

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Impact of new particle formation on the concentrations of aerosol number and cloud condensation nuclei around Beijing

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New particle formation (NPF) is one of the most important processes to determine the concentrations of aerosol number (condensation nucleus, CN) and cloud condensation nuclei (CCN) in the atmosphere. In this study, we developed NPF-explicit WRF-chem model with 20 aerosol size bins from 1 nm to 10 μm and with activation-type nucleation parameterization, which was recently suggested. This model was applied to Beijing region for the periods during the CARE-Beijing 2006 campaign conducted in August and September 2006.

Model calculations reproduced the timing of NPF (21 days out of 26 days measurement available) and the rapid growth of nucleated particles (NP) up to several-tens nanometers reasonably. NPF was mostly observed and calculated in "sweeping" periods when 0-3 days after the cold front passages with the inflow of clean air from the north, while there were few NPF events in "stagnant" periods when Beijing region was influenced from high-pressure system with the accumulation of trace gases and aerosols. The difference of NPF frequency between sweeping and stagnant periods could be explained by the balance of growth rate and condensation and coagulation sink of NP. This result suggests that once the reasonable nucleation rates of cluster formation at 1 nm were given, model calculations can represent the timing of NPF (contrast of "NPF" and "no-NPF" days) and further growth up to several-tens nanometers through the theoretical calculations of condensation and coagulation processes.

The contribution of NPF to the CN concentrations larger than 10 nm (CN10) was estimated to be 20% in Beijing in period average. This contribution became maximum in the noontime (12-16 LT): 73% in NPF days and 17% in no-NPF days, respectively. The impact of NPF on CCN concentrations was dependent on supersaturations (S): CCN concentrations were increased (by 100-200% in maximum) at higher supersaturations ($S > 0.2\%$) but decreased (by 50% in maximum) at lower supersaturations ($S < 0.1\%$) by NPF. This is likely because NPF suppresses the increases in size and hygroscopicity of pre-existing particles through the competition of condensable gases between smaller secondary particles and larger pre-existing particles.

Sensitivity calculations were also conducted with the reduction of primary aerosol emissions (black carbon and primary organic aerosol). We will show the sensitivity of CN and CCN concentrations to primary aerosol emissions in the presentation.

Keywords: New particle formation, Aerosol number concentration, Cloud condensation nuclei, Regional three-dimensional model, Mega city

AAS021-15

Room:102

Time:May 23 12:15-12:30

Comparison of aerosols among surface measurements, CALIOP data, and the SPRINTARS model(I): Sources of dust at Phimai

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For atmospheric aerosols measured at Phimai, Thailand, comparison of chemical and optical properties was performed between the surface measurements and the SPRINTARS model in the previous report at Japan Geoscience Union Meeting 2010. According to the analysis by the field study, dust particles were transported from East China and Indochina, in the early- and the late-dry season, respectively. In contrast, the dust concentration in wet season was usually low compared with that in the dry season, while high concentration of dust was measured, comparable to that in the dry season due to transport of the edge of dust storms in East Asia. The purpose of this study is to clarify if the high dust particles were caused by the local sources or by long range transport. The case study on the episode of high dust concentration during 17-20 June 2008 was made, by comparing the surface data with CALIOP data (<http://www-calipso.larc.nasa.gov/data/BROWSE/production/V3-01/>), NIES RIDAR data(<http://www-lidar.nies.go.jp/Phimai/archives/>), and the result of the SPRINTARS model(<http://sprintars.riam.kyushu-u.ac.jp/archivej.html>). The CALIOP data showed that high dusts were measured during 14-20 June 2008, from Saudi Arabia to Somalia, from Pakistan to Arabian Sea, from Bangladesh to the Bay of Bengal, and the maximum vertical height of the dust layer reached up to 7 km. According to the RIDAR data, high dust aerosols were observed up to the height of 2-3km during 15-17 June 2008 (missing data from 18 June). Furthermore, the backward trajectory analysis by NOAA HYSPLIT MODEL (<http://www.arl.noaa.gov/ready/hysplit4.html>) showed that the air masses arrived at Phimai on 17-20 June 2008, was transported in the lower troposphere from over the Bay of Bengal a few days after, and in the layer of 2-4km height from the east coast of north Africa one week after. On the other hand, the SPRINTARS model, a high dust layer existed up to the height of 2-3km at least for a week from 14 June 2008, spreading horizontally from the eastern part of North Africa to the Bay of Bengal through west Asia, India, and the edge of which reached Indochina. All these data strongly suggest that the high dust particles at Phimai in the wet season could be caused by the long range transport of dust generated in the desert areas of west Asia, in addition to the local dust.

Keywords: atmospheric aerosol, dust, CALIOP, LIDAR, long range transport, SPRINTARS model

AAS021-16

Room:102

Time:May 23 12:30-12:45

Simulating black carbon at Syowa station, Antarctica: long-range transport from various source regions

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This study evaluates long-range transport of black carbon (BC) to the Antarctic region in detail with a chemistry and aerosol coupled global climate model CHASER-SPRINTARS. BC, also called soot, is aerosol species emitted from fossil fuel combustion and biomass burning. BC is one of the most important air pollutants and also causes significant climate impacts with absorbing sunlight and melting snow/glacier. In this study, data derived from the continuous observation at Syowa station (69.0°S, 39.6°E) are used for a representative mass concentration of BC in the Antarctic region. It is found that the current version of the model tends to underestimate BC concentration all year round especially in polar region. This suggests that BC long-range transport process may not be simulated properly in the model, probably due to the model uncertainties in surface emission process and wet deposition process associated with precipitation. To reduce the model underestimation of BC in the Antarctic region, we perform several sensitivity experiments for improving reproducibility of BC long-range transport in the model. In the experiments we reduce the activity as CCN (cloud condensation nucleus) for hydrophobic BC and/or increase the fraction of BC in external mixing for surface emissions (the emission ratio of hydrophobic BC to total BC) to larger. The sensitivity experiments reproduce the observed BC level at Syowa station, but fail to reproduce the observed seasonal cycle of BC with winter high and summer low. Previous studies have suggested that aging process which changes hydrophobic BC to hydrophilic by coating it with water soluble species like sulfate during transport may play an important role in seasonal cycle of BC in remote regions. In this study, we examine the impacts of such aging process on BC in the Antarctic region, newly introducing an aging scheme for BC in the model. As a result, the model with aging process successfully reproduces the seasonal cycle of BC as well as the concentration level at the Syowa station. These sensitivity experiments reveal that long-range transport and subsequent concentrations of BC in remote areas are largely controlled not only by atmospheric transport, but also by BC mixing state and wet-deposition with precipitation and aging effect. In this study, a tagged tracer experiment is also conducted to estimate source region and transport pathway for BC at the Syowa station, Antarctica. The experiment indicates that about 50% of annual mean BC at the Syowa station comes from South America with ~20% from South Africa and 15-20% from Australia. The model reveals two patterns of transport pathway. One is the case that BC is transported to the Syowa station in the lower troposphere (below 3 km altitude). The other case is that BC is first lifted up to the tropopause regions over source region and is transported toward Antarctica via the upper troposphere and lower stratosphere and eventually reaches to the Syowa station associated with Katabatic winds.

Keywords: Black Carbon, Antarctic, Long-range Transport, Chemical Transport Model

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AAS021-17

Room:102

Time:May 23 14:15-14:30

Process study of volatile organic compounds in surface seawater using PTR-MS

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We developed an equilibrator inlet-proton transfer reaction-mass spectrometry (EI-PTR-MS) method for fast detection of dimethyl sulfide (DMS) and volatile organic compounds (VOCs) dissolved in seawater. Dissolved DMS and VOCs extracted by bubbling pure nitrogen through the sample were continuously directed to the PTR-MS. DMS and several other VOCs reached equilibrium with an overall response time of minutes. The detection limit for DMS was 50 pmol L⁻¹ at 5-s integration. The EI-PTR-MS instrument was deployed during a research cruise in the western North Pacific Ocean. For DMS and VOCs, comparison of the EI-PTR-MS results with results obtained by means of membrane tube equilibrator-gas chromatography/mass spectrometry agreed reasonably well on average. EI-PTR-MS captured temporal variations of dissolved DMS and VOCs concentrations, including elevated peaks associated with patches of high biogenic activity. These results demonstrate that EI-PTR-MS was effective for highly time-resolved measurements of DMS and VOCs in the open ocean. Further measurements will improve our understanding of the biogeochemical mechanisms of the production, consumption, and distribution of DMS and VOCs in the ocean surface.

Keywords: volatile organic compounds, DMS, surface seawater, PTR-MS, SOLAS, biogeochemistry

AAS021-18

Room:102

Time:May 23 14:30-14:45

Eight-component retrievals from ground-based MAX-DOAS observations

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We attempt for the first time to retrieve lower-tropospheric vertical profile information for 8 quantities from ground-based Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) observations. The components retrieved are the aerosol extinction coefficients at two wavelengths, 357 and 476 nm, and NO₂, HCHO, CHOCHO, H₂O, SO₂, and O₃ volume mixing ratios. A Japanese MAX-DOAS profile retrieval algorithm, version 1 (JM1), is applied to observations performed at Cabauw, the Netherlands (51.97N, 4.93E), in June-July 2009 during the Cabauw Intercomparison campaign of Nitrogen Dioxide measuring Instruments (CINDI). Of the retrieved profiles, we focus here on the lowest-layer data (mean values at altitudes 0-1 km), where the sensitivity is usually highest owing to the longest light path. In support of the capability of the multi-component retrievals, we find reasonable overall agreement with independent data sets, including a regional chemical transport model (CHIMERE) and in situ observations performed at the 3- and 200-m height levels of the tall tower in Cabauw. Plumes of enhanced HCHO and SO₂ were likely affected by biogenic and ship emissions, respectively, and an improvement in their emission strengths is suggested for better agreement between CHIMERE simulations and MAX-DOAS observations. Analysis of air mass factors indicates that the horizontal spatial representativeness of MAX-DOAS observations is about 3-15 km (depending mainly on aerosol extinction), comparable to or better than the spatial resolution of current UV-visible satellite observations and model calculations. These demonstrate that MAX-DOAS provides multi-component data useful for the evaluation of satellite observations and model calculations and can play an important role in bridging different data sets having different spatial resolutions.

Keywords: MAX-DOAS, retrieval, multi-component, aerosol, ozone

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AAS021-19

Room:102

Time:May 23 14:45-15:00

Regional O₃ trend and its chemical linearity in recent anthropogenic emissions change over East Asia

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Regional O₃ response to perturbations of Chinese anthropogenic emissions is investigated using the brute force method by a regional chemical transport model. Both springtime and summertime ozone responses are unlikely to show nonlinearly in the emissions perturbation of 70%–200% over East Asia. Observed NO₂ in the east central China in 2003–2008 is ranging within a factor of 0.82–1.35 of the 2004 level, and that explains recent O₃ change stays within the linear O₃ response range. As for observed O₃ at Japanese remote sites, the O₃ response is 11.7 ppbv with respect to a 100% increase at the east central China from NO₂ level in 2004. The simulated relationship between O₃ and NO₂ shows 5.9 ppbv of O₃ growth is caused by doubled NO₂ from the 2004 level.

Keywords: tropospheric O₃, chemical linearity, emissions, East Asia

AAS021-20

Room:102

Time:May 23 15:00-15:15

CO emissions from biomass burning in Southeast Asia in the 2006 El Nino year: Shipboard and AIRS satellite observations

Hideki Nara^{1*}, Hiroshi Tanimoto¹, Yukihiro Nojiri¹, Hitoshi Mukai¹, Jiye Zeng¹, Yasunori Tohjima¹, Toshinobu Machida¹

¹NIES

[Introduction]

During the dry season (October-November) in the 2006 ENSO year, we observed substantial CO enhancement over the western tropical Pacific by shipboard observations routinely operated between Japan and Australia/New Zealand. In this presentation, we present evidence of CO pollution episodes over the tropical Pacific due to intensive biomass fires in Southeast Asia and north Australia during 2006 El Nino year. We discuss locations of CO emissions from fires followed by long-range transport by combination of in-situ ship measurements, Atmospheric Infrared Sounder (AIRS) satellite observations, and Lagrangian particle dispersion model (FLEXPART).

[Method]

Beginning in November 2005, continuous monitoring of atmospheric trace gases has been initiated by using a commercial cargo vessel, M/V Transfuture 5 (TF5) (owned by Toyofuji Shipping Co. Ltd.). The ship takes regular service among ports of Japan, Australia and New Zealand with the 6 weeks interval, covering subtropical latitudinal region over North and South Pacific. The shipping route of TF5 is shown in Figure. Automated instruments were installed into an observation room for in situ measurements of CO, CO₂ and O₃.

[Result & Discussion]

Abnormal enhancements in CO were observed between 15N and the Equator during the southbound voyage (Episode 1), and around the Equator (Episode 2) during the northbound voyage, which had large impact on the seasonal variations of CO in the tropical Pacific. During Episode 1, AIRS satellite images and Global Fire Emissions Database version 2.1 (GFEDv2.1) suggested that the CO plume originated from biomass burning in Borneo and Sumatra followed by long-range transport to the tropical Pacific region. The amplitude of observed CO enhancement during Episode 2 was much smaller than that during Episode 1. Simulations by FLEXPART showed well consistent results with our analysis, which provides a reasonable interpretation of the data, complimenting in situ and satellite observations.

Correlations of CO to CO₂ and of O₃ to CO observed during two episodes were also examined. Scatter plots of CO versus CO₂ during Episode 1 showed significant correlation ($R^2 = 0.60$) with a steep dCO/dCO_2 slope (171 ± 31 ppbv/ppmv at the 95% confidence interval). The dCO/dCO_2 ratio observed was higher than previously reported for savanna and grassland (63 ± 20 ppbv/ppmv), tropical forest (103 ± 21 ppbv/ppmv), and slightly higher than Indonesian peatland fires 142.7 ppbv/ppmv, indicating that Indonesian fires including peat soil burning are a dominant factor during Episode 1. Comparison between the dCO/dCO_2 ratio and CO/CO₂ emission ratios from GFEDv2.1 suggests the uncertainty in CO emissions of GFEDv2.1 in Southeast Asia region associated with peatland fires. Significant O₃-versus-CO correlation was observed only for Episode 1 ($R^2 = 0.68$). The dO_3/dCO ratio (0.05 ± 0.01 ppbv/ppbv) was considerably smaller than values reported in previous observations in this region, which suggests that net O₃ production was not efficient in the burning plumes transported in the lower troposphere over the western tropical north Pacific. The reason for low dO_3/dCO ratio is not clear, but is likely associated with combustion properties of the peat and/or meteorological condition during the transport, both of which are specific to Southeast Asia region. Further research is needed to understand the low dO_3/dCO ratio in the long-range transport of the burning plumes over western tropical north Pacific in the lower troposphere.

Keywords: Carbon monoxide, Voluntary observing ship, AIRS, Southeast Asia, Biomass burning

AAS021-21

Room:102

Time:May 23 15:15-15:30

Development of the long-term mass transport model and verification by Chernobyl accident and Sakurajima volcano

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In those days, environmental problems are taken up as the societal issues. Air pollution is one of them. The air pollution cases have been confirmed all over the world. There are some cases that require the long-term prediction and behavior grasp. For example, the radionuclide pollution in Chernobyl and the volcanic gas exposure in the surrounding area. To predict the air pollution and grasp the condition of the air, many models have been studied. Gaussian plume model is the most used among them. However, most models including the model don't assume the long-term prediction.

In this study, we propose the model suited for the long-term prediction, which have not studied enough, and verify the applicability of our model by fitting with measured data. The long-term model in this study is constructed by the governing equation based on the advection equation, and the approximate analytical solution is derived from the mathematical method. The governing equation is the following equation.

$$dC/dt + \{(v_x)d/dx + (v_y)d/dy\}C + I_{env}C + I_{decay}C = P(x, y, t)$$

where C is the concentration in the air, t is the days since the standard time, and v is each forward advection velocity by wind. I_{env} is a local environmental removal rate, that is, the sum of reaction rate by local environmental kinetics such as chemical reactions of the nuclide with soil, permeation into soil, vegetation uptake, water run off, etc. I_{decay} is the physical decay.

$P(x, y, t)$ is the term expressing the emission of pollutant, and the analytical solution differs according to the definition of $P(x, y, t)$.

In this study, the pollution types were divided into four types. As the emission form, the instantaneous emission and the continuous emission were assumed. As the emission source type, the point source and the plane source were assumed.

Fitting with measured data, P was defined according to the cases and the analytical solution was derived in each cases. Radionuclide diffusion derived from Chernobyl Nuclear Power Plant accident was assumed as the instantaneous emission at the point source. In addition, SO_2 diffusion derived from Sakurajima Volcano was assumed as the continuous emission at the point source. And derivation of the model (the analytical solution) was assumed about the both cases.

In case of the instantaneous emission at the point source, P was defined as the following equation.

$$P = \delta(x)\delta(y)\delta(t).$$

and the model was derived. Here, $\delta(x)$ is delta function. As the result, the model was derived as

$$C(t) = A \exp(-I_{decay} t) t^{-a}.$$

A and a are fitting parameters.

In case of the continuous emission at the point source, P was defined as the following equation.

$$P = \delta(x)\delta(y)\{P_1\delta(t_1) + \dots + P_n\delta(t_n)\}.$$

P was defined as the sum of delta functions according to times of explosion in this case. The model became the equation including the Fourier transform. So the calculation cannot complete by calculating parameters, and the calculation result was calculated by Mathematica.

As a result, the fitting of 4 radionuclides from Chernobyl Nuclear Power Plant accident was successful at 21 observation points within 40 km. The fitting of period was from 3000 days to 5000 days. The fitting was also showed that calculated parameters A and a has a positive correlation totally but a feature of a positive correlation varied by each radionuclide. Moreover, the fitting of SO_2 from Sakurajima Volcano was also successful with the data of Arimura station which can be most affected by Sakurajima Volcano. The period of fitting was from 2002 to 2008. This result showed that the model can calculate the annual concentration within ± 0.005 error.

The above results about two pollution cases shows that the model we proposed was suitable to the long-term prediction of concentration in the air. It is assumed our model can be used in case of the more complicated case such as the continuous emission at the plane source.

Keywords: long-term mass transport model, Chernobyl, Sakurajima volcano, air pollution, dispersion predicting

AAS021-22

Room:102

Time:May 23 15:30-15:45

Variations of atmospheric radon-222 at Rishiri Island, Japan and traced fetch regions

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Measurements of atmospheric ²²²Rn were made on Rishiri Island (45.1°N, 141.2°E) over the period from December 2008 to December 2010, in order to discuss the fetch regions affecting atmospheric ²²²Rn concentration and some other gases related to climate change. Atmospheric ²²²Rn data showed variability of a diurnal, synoptic, and seasonal time scales, which will allow us to evaluate transport and mixing schemes of atmospheric and chemical transport models.

Atmospheric ²²²Rn concentration indicated a clear diurnal variation in summer, which was characterized of a maximum appearing before the dawn and a minimum in the afternoon, with amplitude of 0.64 Bq m⁻³. This could be caused by the accumulation of ²²²Rn emitted from the soil (Rishiri Island) in the stable nocturnal boundary layer, and vertical mixing of surface air with upper air due to convection (Zahorowski *et al.*, 2008). The amplitude of diurnal variation is relatively lower than that of the synoptic influence. No clear diurnal variation occurred during the remaining seasons.

Atmospheric ²²²Rn concentration in daytime, selected to remove effects of local source on ²²²Rn concentration, showed a broad minimum in winter and a maximum in summer, with an accompanying significant short-term variability. In February monthly mean of atmospheric ²²²Rn concentration was 3.23 Bq m⁻³, and in July 0.95 Bq m⁻³. In Rishiri Island, short-term variations corresponding to the synoptic influence were large in February as compared with those in July. The amplitude of the seasonal variation (2.28 Bq m⁻³) was somewhat larger than that of Sado Island located in the Sea of Japan. At Sado Island, maximum ²²²Rn concentration occurred in winter and minimum in summer, which was the pattern attributable to the onshore-offshore pattern of the Asian monsoons (Chambers *et al.*, 2009).

Backward trajectory analysis of air fetch regions was conducted using extremely low and high radon events. In the annual basis, most (76.7%) of high radon events was observed in winter, of which the air masses originated predominately from 40°N to 60°N of Eurasian continent. 41.7% of low radon events were observed in summer, of which air masses usually originated from the relatively lower latitude of the western North Pacific, and 48.3% of low radon events were observed in spring, most in May. In May, back trajectory analysis showed a pattern that the origin of air masses was in the west North Pacific, and air mass moved westward en route the southernmost of Sea of Okhotsk to the RIO.

Keywords: atmospheric tracer, back trajectory analysis, fetch regions, radon

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AAS021-23

Room:102

Time:May 23 15:45-16:00

Night time radical chemistry observed by SMILES

YASUKO KASAI¹, Philippe Baron^{1*}, Hideo Sagawa¹, Donal Murtagh², Joachim Urban², BrO team in SMILES Algorithm team¹

¹NICT, ²Chalmers Institute of Technology

A new generation of super-sensitive submillimeter-wave receivers, employing SIS (Superconductor-Insulator-Superconductor) technology, will provide new opportunities for precise remote sensing observation of minor constituents in the atmosphere. SMILES had been launched at 11/09/2009, and installed on the Japanese Experiment Module (JEM) in the International Space Station (ISS). SMILES is a collaboration project between NICT and JAXA.

Mission objectives of SMILES are:

- i) Space demonstration of super-sensitive SIS mixer and 4-K mechanical cooler technology
- ii) Demonstration of super-sensitive global observation of atmospheric minor constituents

JEM/SMILES observes the atmospheric species such as O₃, H₃₅Cl, H₃₇Cl, ClO, HO₂, BrO, HOCl, HOBr, HNO₃, CH₃CN, Ozone isotope species, H₂O, and Ice Cloud with the precisions in a few to several tens percents. We will present the diurnal variation of the minor radical such as BrO, HO₂.

Keywords: SMILES, BrO

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AAS021-24

Room:102

Time:May 23 16:00-16:15

Diurnal variation of ClO observed with SMILES

Yu Onodera^{1*}, Tomohiro Sato³, Philippe Baron², YASUKO KASAI², Eric Dupuy², Kazuyuki Kita¹, Joachim Urban⁴, Donal Murtagh⁴, Nathaniel Livesey⁵

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A new generation of super-sensitive submillimeter-wave receivers, employing SIS (Superconductor-Insulator- Superconductor) technology, will provide new opportunities for precise remote sensing observation of minor constituents in the atmosphere. SMILES had been launched at 11/09/2009, and installed on the Japanese Experiment Module (JEM) in the International Space Station (ISS). SMILES is a collaboration project between NICT and JAXA.

SMILES has observed the diurnal observation of ClO, HOCl, HO₂, and HCl from October 12, 2009 to April 21, 2010. We will present 1) the result of the evaluation of ClO data quality, including inter-comparison between other satellites measurements and error analysis, and 2) the first simultaneous observations of ClO, HOCl, HO₂, and HCl.